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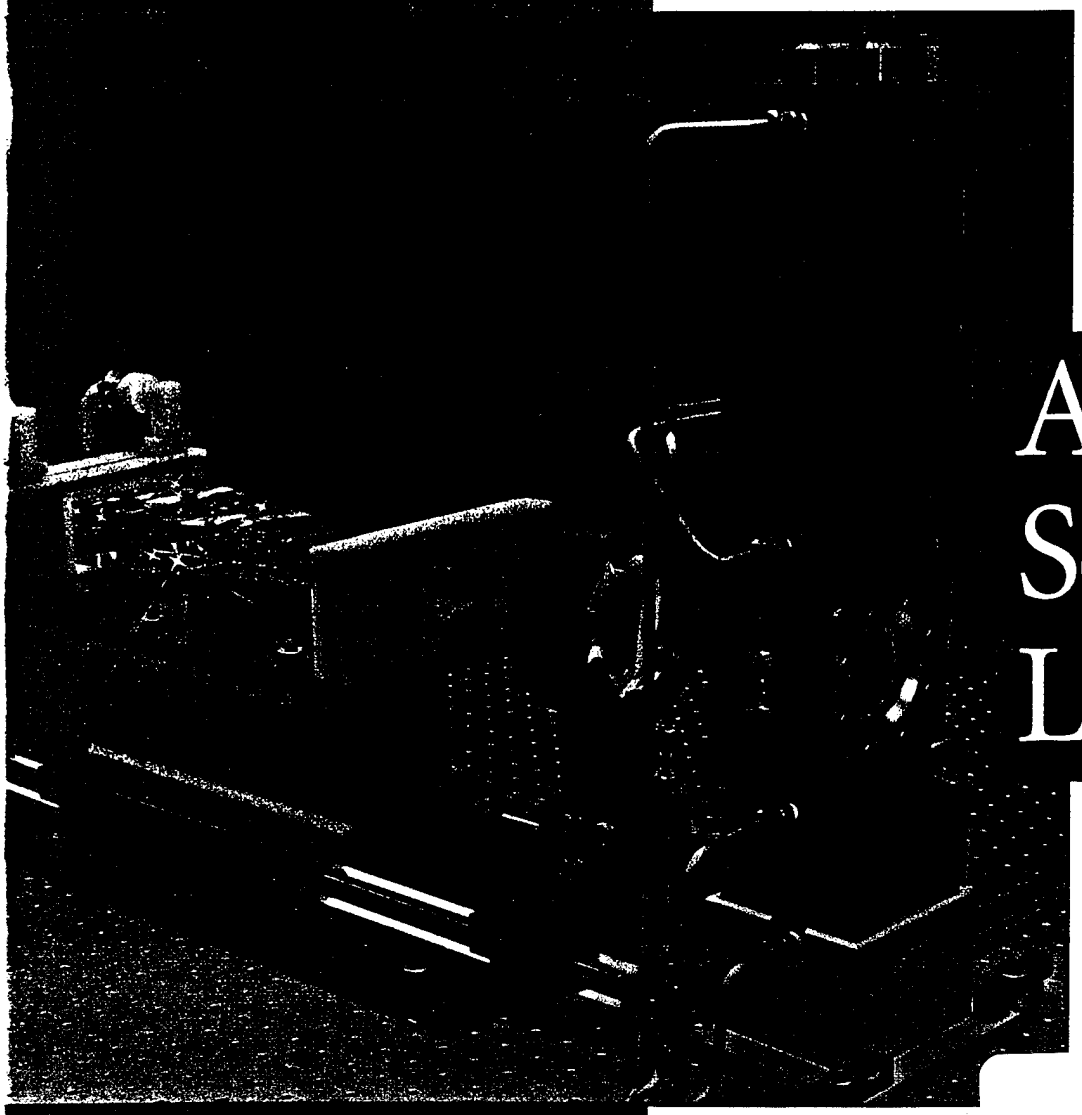

Advanced Solid State Lasers

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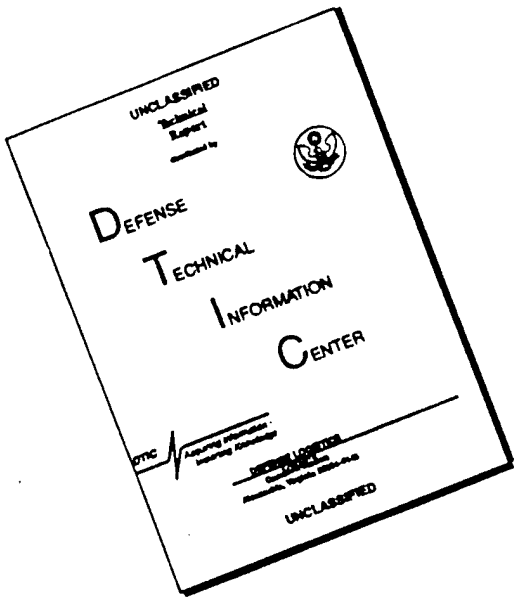

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Stimulated emission without cavity in powders and single crystals of Nd doped materials

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Abstract

Short (>300 ps) pulses of stimulated emission were found from powders of $NdAl_3(BO_3)_4$, $NdSc_3(BO_3)_4$, and $Nd:Sr_5(PO_4)_3F$ laser crystals under 532 nm and 805 nm excitation. Study of stimulated emission in the mixture of two powders has shown that different components influenced each other. The main features of experimentally observed stimulated emission are described with a simple model accounting for $^4F_{3/2}$ excited state concentration and emission energy density.

Key Words

Rare earth and transition metal solid-state lasers (432); Laser theory (86); Scattering by particles (2157); Microstructure devises (454).

Introduction

In 1986 Markushev *et al.* [1] showed that a powder of $Na_5La_{1-x}Nd_x(MoO_4)_4$ at liquid nitrogen temperature pumped at λ=575-590 nm exhibited a laser-like behavior without an external cavity. After some threshold pump energy, the Nd emission spectrum narrowed to a single intense line and short emission pulses appeared in response to a 30 ns Q-switched laser pulse. In succeeding papers by the same group [2-4] the spectral and temporal behaviors of the emission pulses were studied in more detail in various Nd doped crystals. Laser-like behavior of Nd emission in polycrystals of $Nd_xLa_{1-x}P_5O_{14}$ and powders of $(NdCl_3:6H_2O)$ was first demonstrated in Ref. [5] at room temperature. In the same work emission pulses above the threshold were found to be of low coherence.

The short-spike formation and narrowing of the emission spectrum is thought to be due to collective behavior of many particles, where emission is amplified in the gain medium. The diffusion of photons in gain scattering medium was studied by Letokhov in Ref. 6. However, a detailed explanation of the phenomenon is not currently available in the literature.

From the practical point of view, the study of laser-like emission in powders is very interesting because of potential applications of compact, low-cost, and very simply designed lasers based on powders of laser crystals, which do not need any mirrors and adjustment.

Experimental measurements

In the experiment, we studied powders of the $NdAl_3(BO_3)_4$, $NdSc_3(BO_3)_4$, and $Nd(2\%):Sr_5(PO_4)_3F$ (Nd:S-FAP) laser crystals. An average size of the powder particles was approximately equal to 5 μm. The samples were pumped with Q-switched frequency doubled Nd:YAG laser or 805 nm Cr:LiCAF laser.

Experimentally we analyzed spectra and kinetics of Nd emission at low and high pumping densities. All experiments were carried out at room temperature. We found that, after the pump energy exceeds some threshold value, both the kinetics and the spectra of Nd luminescence change very dramatically. Above the threshold, the luminescence spectrum narrows down to a single line, the measured value of the full width at half height (FWHH) equal to 2 Å was limited by the monochromator. Figure 1 shows the transformation of the spectrum in the $NdAl_3(BO_3)_4$ powder with increase of pumping intensity. The spectrally narrow light was emitted in one or several short pulses. In $NdAl_3(BO_3)_4$ the duration of the pulses varied from ≈300 ps to ≈1.3 ns (Fig. 2). The other materials studied demonstrated similar behavior. As is typical for most lasers, the dependence of the stimulated emission intensity from the powder on the pump energy is as presented in Fig. 3.

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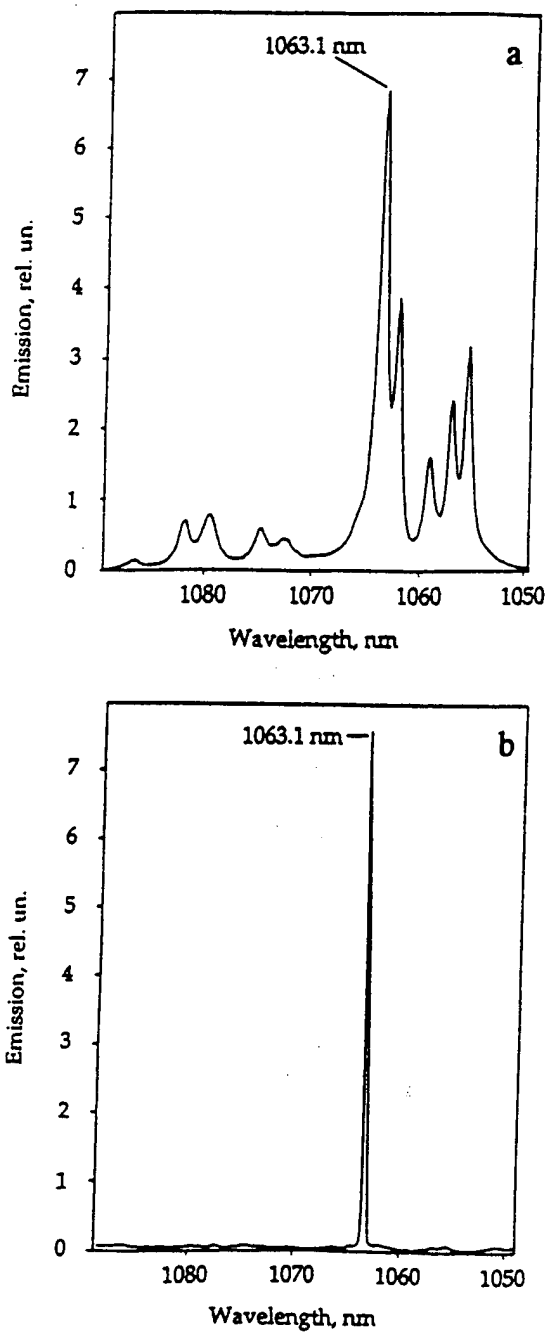

Figure 1. Emission spectrum of $NdAl_3(BO_3)_4$ powder a) below the threshold (≈30 mJ/cm^2) and b) above the threshold (≈240 mJ/cm^2), λ=1063.1 nm.

The threshold energy density, the threshold Nd excited state concentration, and the threshold gain at the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition in the three powder samples studied are summarized in Table 1. The energy levels diagram of Nd^{3+} ions is presented in Fig. 4.

An apparently similar emission behavior we observed under pulsed Ti-sapphire pumping (808 nm, 20 ns) in polished plane-parallel samples of Nd:GVO_4

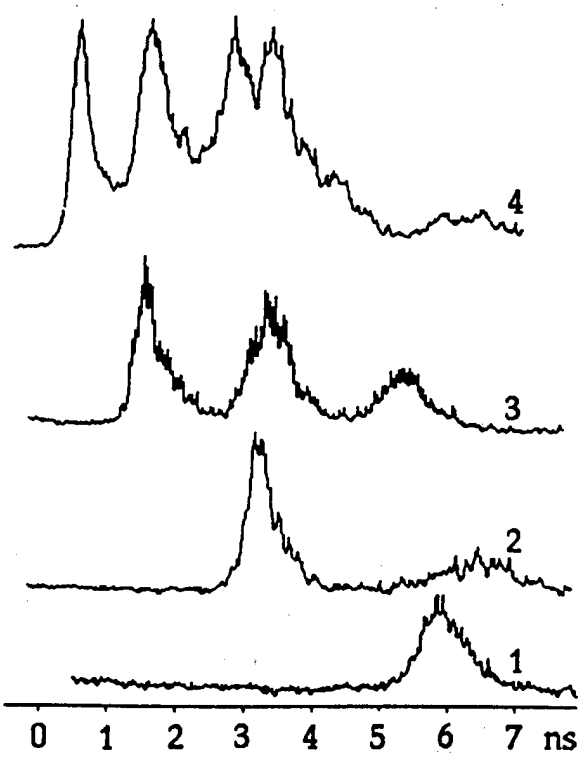

Figure 2. Pulses of stimulated emission from $NdAl_3(BO_3)_4$ powder 1) near the threshold (200 mJ/cm^2), 2) at x=1.6 times threshold energy, 3) x=1.9, 4) x=3.9.

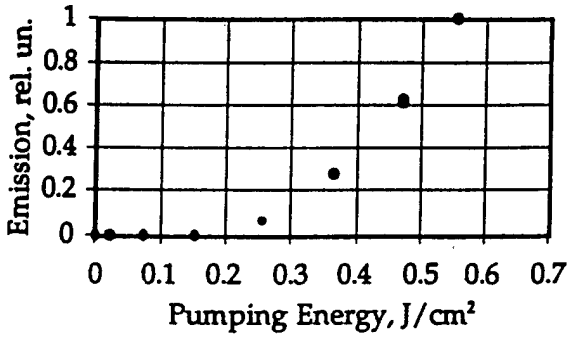

Figure 3. Experimental dependence of the intensity of stimulated emission in $NdAl_3(BO_3)_4$ on the pump energy, λ=1063.1 nm.

(Nd=0.9%, 2.8%) and $Nd_xLa_{1-x}Sc_3(BO_3)_4$ (x=0.1, 0.25). Short and very intense emission spikes were observed in both materials when Ti-sapphire laser was tuned to the maximum of Nd absorption. In both crystals the ratio of the spike intensity to the intensity of the succeeding "regular" luminescence was approximately equal to 10^4. Above the threshold, the dependence of spike intensity on pump energy was practically linear.

	$NdAl_3(BO_3)_4$	$NdSc_3(BO_3)_4$/ $Nd_xLa_{1-x}Sc_3(BO_3)_4$	Nd:S-FAP
Nd concentration in 100% doped sample	5.3×10^{21} cm^{-3}, Ref. [7]	5.1×10^{21} cm^{-3} in $NdSc_3(BO_3)_4$., Refs. [8,9]	1.68×10^{22} cm^{-3}, Ref. [10]
$^4F_{3/2}$ life-time	20 μs, Ref. [7]	Nd(1-10%): $LaSc_3(BO_3)_4$ 118 μs $NdSc_3(BO_3)_4$ 24 μs, Refs. [8,9]	298 μs, Ref. [10]
Wavelength of the maximum emission cross section	≈1.063 μm Ref. [11]	1.0615 μm, (our measurements)	1.059 μm, Refs. [10,12]
$^4F_{3/2}$–$^4I_{11/2}$ emission cross section	10×10^{-19} cm^2, Ref. [7]	$Nd_{0.1}La_{0.9}Sc_3(BO_3)_4$ Ellx, 13×10^{-20} cm^2, Elly, 9×10^{-20} cm^2, Ellz, 5×10^{-20} cm^2, Ref. [8], averaged over different polarizations: 9×10^{-20} cm^2	Ellc, 5.4×10^{-19} cm^2, E⊥c, 2.4×10^{-19} cm^2 Refs. [10,12], averaged over different polarizations: 3.9×10^{-19} cm^2
Absorption cross section at the pump wavelength	Ellc 3.3×10^{-21} cm^2, Ellc, 2.6×10^{-21} cm^2 (λ=532 nm); Ref. [13]	Ellx, 5.1×10^{-21} cm^2, Elly, 4.2×10^{-21} cm^2, Ellz, 2.4×10^{-21} cm^2 (λ=532 nm); Ref. [14]	Ellc, 2.26×10^{-19} cm^2 E⊥c, 0.7×10^{-19} cm^2 (λ=805 nm); Ref. [10]
Absorption coefficient of the powder (averaged over different polarizations)	≈15 cm^{-1} (λ=532 nm, 100%Nd)	≈20 cm^{-1} (λ=532 nm, 100% Nd)	≈55 cm^{-1} (λ=805 nm, 2% Nd)
Threshold pump density (in the powder)	200 mJ/cm^2 (λ=532 nm)	560 mJ/cm^2 (λ=532 nm)	170 mJ/cm^2 (λ=805 nm)
Threshold Nd excited state concentration	7.5×10^{18} cm^{-3}	2.8×10^{19} cm^{-3}	3.64×10^{19} cm^{-3}
Threshold gain	≈7.5 cm^{-1}	≈2.5 cm^{-1}	≈15.7 cm^{-1}

Table 1. Spectroscopic data on $NdAl_3(BO_3)_4$, $Nd_xLa_{1-x}Sc_3(BO_3)_4$, and Nd:S-FAP laser crystals. (In $Nd_xLa_{1-x}Sc_3(BO_3)_4$ the most of data are available on low Nd doped crystals. The change of the crystal symmetry at >50% Nd concentration may influence some of spectroscopic parameters.)

Similar laser-like effects in the emission from small single crystals of $NdAl_3(BO_3)_4$ were reported in Ref. [7].

To compare the behavior of spiked emission from powders to that from single crystals, we studied polished and unpolished plates of the Nd(2%):S-FAP single crystals, and small (≈1mm^3) single crystals of $NdAl_3(BO_3)_4$ and $NdSc_3(BO_3)_4$ (Table 2); we also studied a thin layer of $NdAl_3(BO_3)_4$ powder. As follows from Table 2, three factors help stimulated emission to occur: 1) appreciably large pumped volume, 2) preparation of the material in the powder form (scattering), and 3) polished plane-parallel surfaces in the bulk crystals (feedback). (Note, that the last result is different from that of Ref. [15], where the reflections off the cell walls did not decrease but increased the threshold in the dye+powder gain scattering medium).

To determine the portion of the energy stored at the level $^4F_{3/2}$ went to the stimulated emission channel, we studied a) the kinetics of luminescence at the $^4F_{3/2}\rightarrow{}^4I_{9/2}$ transition during the stimulated emission pulses, b)

	$NdAl_3(BO_3)_4$ λ_{pump}=532 nm		$NdSc_3(BO_3)_4$ λ_{pump}=532 nm	Nd:S-FAP λ_{pump}=805 nm	
Powders	V≥1 mm^3	200 mJ/cm^2	560 mJ/cm^2 (V≥1 mm^3)	170 mJ/cm^2 (V≥1 mm^3)	
	thin monolayer of powder	at ≤1 J/cm^2 threshold was not achieved	–	–	
Single crystals	V≈1 mm^3	600 mJ/cm^2	–	8 mm polished plate	625 mJ/cm^2
				1.5 mm polished plate	920 mJ/cm^2
				0.8 mm unpolished plate	1080 mJ/cm^2

Table 2. Thresholds of the stimulated emission in the powders and single crystals of $NdAl_3(BO_3)_4$, $Nd_xLa_{1-x}Sc_3(BO_3)_4$, and Nd:S-FAP laser crystals.

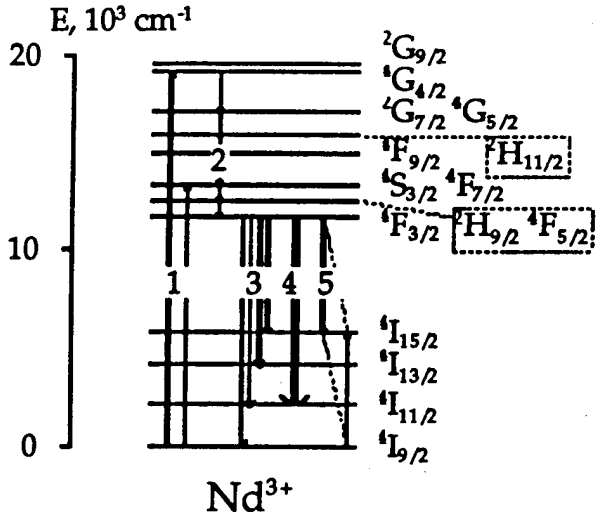

Figure 4. Nd^{3+} energy levels diagram, excitation and relaxation processes in Nd^{3+}: 1) - pumping, 2) - multiphonon relaxation populating the metastable level $^4F_{3/2}$, 3) radiation and multiphonon relaxation of the level $^4F_{3/2}$, 4) stimulated emission at the transition $^4F_{3/2} \rightarrow {}^4I_{9/2}$, 5) cross relaxation.

analyzed the dependence of the peak $^4F_{3/2} \rightarrow {}^4I_{9/2}$ emission *vs* pump energy, and c) compared the area under the stimulated emission pulses and residual spontaneous emission kinetics from the level $^4F_{3/2}$. Combining the results of the three measurements above, we estimated the quantum yield of stimulated emission to be rather small, ≈0.2%, at the pump energy two times greater than the threshold energy. However, accounting for the small pulse duration of the stimulated emission, the peak power of the stimulated emission was comparable to that of the pump pulses.

We then turned to the study of mixtures of the two powders. We tried ≈1/6 $NdAl_3(BO_3)_4$ and ≈ 5/6 $NdSc_3(BO_3)_4$, by volume. In the mixtures, the maximum strong luminescence line of $NdAl_3(BO_3)_4$

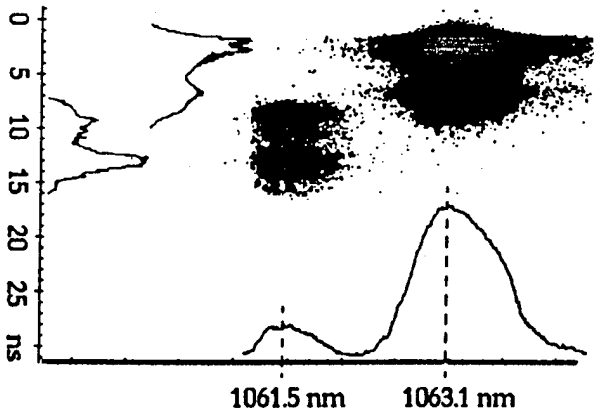

Figure 5. Image of stimulated emission from the mixture of ≈1/6 $NdAl_3(BO_3)_4$ and ≈ 5/6 $NdSc_3(BO_3)_4$ as it appears on the screen of streak camera connected to monochromator

(λ=1063.1 nm) was practically not seen under the relatively wide line of $NdSc_3(BO_3)_4$. Above the threshold, however, two narrow emission lines (1063.1 nm and 1061.5 nm) appeared. Their thresholds and relative strengths were strongly dependent on very small variations of concentrations of the components.

Combining the streak camera with the monochromator, we obtained a three-dimensional picture (wavelength-time-intensity) of the stimulated emission in the mixture of two powders, Fig. 5. When the pump energy was above the threshold required for both lines, several short pulses of emission appeared at 1063.1 nm, after that emission jumped to 1061.5 nm which gave one or several short pulses, depending on the pump energy.

In a one-component medium the first pulse in the series was always the strongest one, Fig. 2. In contrast, in the mixture of two powders (see Fig. 5), where the first pulse in 1061.5 nm series coincided in time with the last pulse in 1063.1 nm series, the first 1061.5 nm pulse was strongly damped and weaker than succeeding

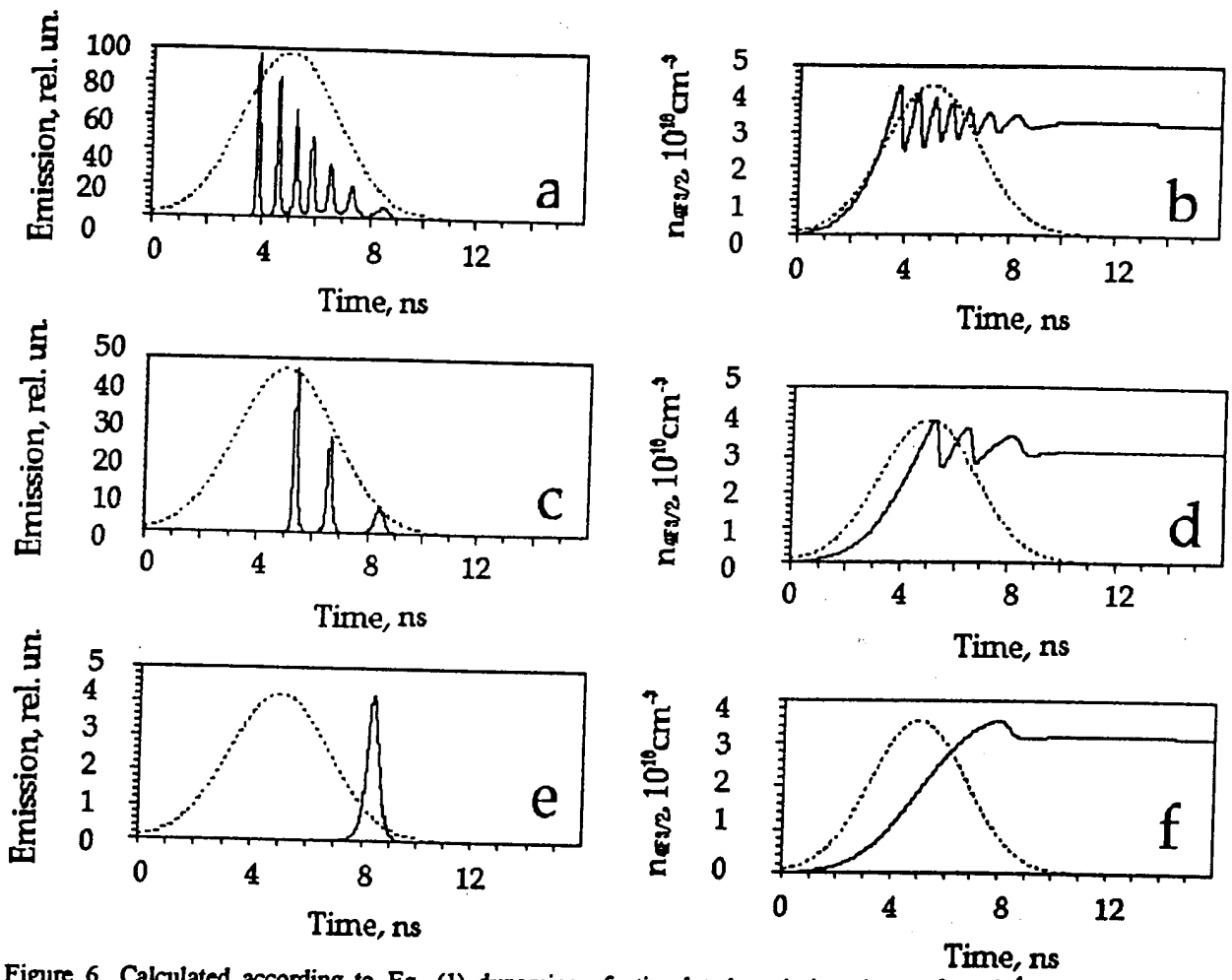

Figure 6. Calculated according to Eq. (1) dynamics of stimulated emission (a, c, f) and $^4F_{3/2}$ excited state concentration (b, d, e) in $NdAl_3(BO_3)_4$. Pumping density: 1000 mJ/cm^2 in a,b, 400 mJ/cm^2 in c,d, and 200 mJ/cm^2 (the threshold) in f,e. Dashed line - pump pulse.

pulses of the same series. From this observation we conclude that in the mixture individual components do not operate independently. This also implies that in one-component medium many particles behave collectively to produce stimulated emission pulses.

Comparison of experiment and theory. Discussion

The threshold behavior of stimulated emission and the short emission pulses we describe by a simple model accounting for the Nd excited state concentration, n, and emission energy density, E, in the pumped volume:

$$\frac{dn}{dt} = \frac{K(t)N\sigma_{abs}}{h\nu_{pump}} - \frac{n}{\tau_1} - \beta nN - \frac{E}{h\nu_{em}} c\sigma_{em} n$$
$$\frac{dE}{dt} = -\frac{E}{\tau_2} + \frac{n}{\tau_1} h\nu_{em} + Ec\sigma_{em} n \qquad (1)$$

Here K(t) is the pump power density, N is the ground state concentration of Nd, σ_{abs} is the absorption cross section at the pump wavelength, σ_{em} is the emission cross section at the wavelength of stimulated emission, $h\nu_{pump}$ is the photon energy at the pump wavelength, $h\nu_{em}$ is the photon energy at the emission wavelength, τ_1 is the life-time of the level $^4F_{3/2}$, βN is the rate of cross relaxation, τ_2 is the effective life-time of the photon in the pumped volume, c is the speed of light.

The idea behind Eq. (1) (to watch for dynamics of emission energy in the pumped volume) is close to that of Letokhov [6] who calculated threshold for stimulated emission in the gain scattering medium. Equation (1) is also similar to that for laser relaxation oscillations, where τ_2 has a meaning of the photon life-time in the laser cavity.

In the numerical solution of Eq. (1) we used the spectroscopic parameters close to that in the $NdAl_3(BO_3)_4$ experiment. The only unknown parameter in Eq. (1) was τ_2, the effective photon life-time in the pumped volume. We used it as an adjustable parameter

to fit the experimental energy threshold and found τ_2=10 ps. At n'=1.5 this value of τ_2 corresponds to the average 2 mm photon path in the pumped medium. This seems to be a reasonable value for ≈1 mm pump beam cross section and ≈0.7 mm absorption length (in $NdAl_3(BO_3)_4$) in our experiment.

The calculated dynamics of emission energy density and $^4F_{3/2}$ exited state concentration are shown in Fig. 6. The appearance of calculated emission pulses is very close to that observed experimentally. The analysis of excited state concentration dynamics, n(t), shows that pulses of stimulated emission occur when n exceeds some critical threshold value, practically independent of pump density.

We have shown that the calculated threshold of stimulated emission is inversely proportional to the absorption coefficient at the pump wavelength, $k_{abs}=N\sigma_{abs}$ (pump efficiency), emission cross section, σ_{em}, and escape-time of photon from the pumped volume, τ_2 (photon walk length in the pumped volume). Thus, the threshold is inversely proportional to the small signal amplification along the effective photon path in the pumped volume. This result is in an agreement with our experimental observations. For lasers the described threshold behavior is obvious and have been demonstrated elsewhere.

According to our simple model, preparation of the sample in powder form and polishing of surfaces do not produce any special condition necessary for stimulated emission to occur, but only reduce the threshold of stimulated emission. Short pulses of stimulated emission are predicted (and obtained experimentally) at higher but still reasonable pumping energy in excited crystal without any scattering and reflection feedback.

Summary

Room temperature stimulated emission was found in the powders of $NdAl_3(BO_3)_4$, $NdSc_3(BO_3)_4$, and Nd:SFAP laser crystals under Q-switched laser pumping: when the pump energy exceeded the threshold value, Nd emission spectrum narrowed to a single line, one or several short (>300 ps) emission pulses appeared in response to the pump pulse.

Similar behavior, but at higher thresholds, was found in single crystals of the same materials. However, no stimulated emission was obtained in a monolayer of powder. Thus, appreciably large pumped volume and long paths of emission photons help to reduce the threshold of stimulated emission. It was shown that only small portion of excitation stored at the $^4F_{3/2}$ Nd level goes to the stimulated emission channel.

Study of stimulated emission in the mixture of two powders has shown that different components influence each other, that implies a collective behavior of many emitting particles.

The main features of the experimentally observed short pulsed emission were described with a simple model accounting for the $^4F_{3/2}$ excited state concentration and emission energy density in the pumped volume.

Acknowledgments

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References

1. V. M. Markushev, *et al.*, Sov. J. Quantum Electronics, **16**, pp. 281-283 (1986).
2. V. M. Markushev, *et al.*, Sov. J. Quantum Electron., **20**, pp. 773 - 777 (1990).
3. N. É. Ter-Gabriélyan, *et al.*, Sov. J Quantum Electron., **21**, pp. 840-842 (1991).
4. N. É. Ter-Gabriélyan, *et al.*, Sov. J. Quantum Electron., **21**, pp. 32-33 (1991).
5. C. Gouedard, *et al.*, J. Opt. Soc. Am. B, **10**, pp. 2358-2363.
6. V. S. Letokhov, Soviet Physics JETP, **26**, pp. 835-840 (1968).
7. G. Huber, "Miniature neodymium lasers", in the book Current Topics in Materials Science, Vol. **4**, E. Kaldis, editor, North-Holland, 1980, pp. 1-45.
8. J.-P. Mein, *et al.*, IEEE Journal Quant. Electron., **30**, pp. 913-917 (1994).
9. V. Ostroumov, *et al.*, OSA Proceeding on Advanced Solid-State Lasers, (OSA, Washington, DC 1995), Vol. **24**, pp. 509-513.
10. X. X. Zhang, *et al.*, Appl. Phys. Lett., **64**, pp. 3205-3207 (1994).
11. G. Huber, H. G. Danielmeyer, Appl. Phys., **18**, pp. 77-80 (1979).
12. X. X. Zhang, *et al.*, OSA Proceeding on Advanced Solid-State Lasers, (OSA, Washington, DC 1995), Vol. **24**, pp. 150-152.
13. H.-D. Hattendorf, Dissertation zur Erlangung des Doktorgrades des Fachbereich Physik der Universität Hamburg, Hamburg, 1979, 97 p.
14. J.-P. Meyn, Dissertation zur Erlangung des Doktorgrades des Fachbereich Physik der Universität Hamburg, Hamburg, 1994, 115 p.
15. Balachandran, N. M. Lawandy, Opt. Lett, **20**, pp. 1271-1273 (1995).